Van Hove singularities in doped twisted graphene bilayers studied by scanning tunneling spectroscopy.

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Twisted bilayers (tBL) form a class of graphene based material whose low energy electronic structure can be controlled by a geometric parameter, namely by the rotation angle θ between the graphene layers. For undoped (neutral) systems, this property has been established by a number of theoretical [1,2] and experimental studies [3,4]. For large angles $(\theta > 10^\circ)$, the layers are electronically decoupled and the low energy band structure looks like a simple superposition of the Dirac cones of the individual graphene planes. For smaller angles, a pair of logarithmic divergences in the density of states (DOS) called van Hove singularities (vHs), related to a saddle point in the band structure, develop within 1 eV from the Fermi level [2-4]. Their energies are almost symmetric with respect to the Dirac point and decrease with θ . For even smaller angles ($\theta < 1-2^{\circ}$), flat bands appear at low energy [2], the vHs tend to localize in AA stacked areas [2,3] and additional low energy DOS features related to confinement appear [2]. The twist induced changes in the band structure should be reflected in the physical properties of the bilayers. A rich physics is anticipated in magnetotransport experiments provided the Fermi level E_F can be brought in the vicinity or above the vHs [5], although it has not yet been revealed by the experiments reported so far. Structures in the optical conductivity induced by the presence of the vHs have been reported in a wide energy range depending on the value of θ [6]. Calculations moreover predict that doping could markedly influence the optical properties when E_F reaches the vHs [7]. Owing to the variety of original properties expected upon doping, it is important to determine in a direct way the influence of this parameter on the low energy electronic structure of the twisted graphene layers. This is especially interesting for the doping levels which are accessible using a backgate, typically a few 10^{12} cm⁻², to determine for instance at which angle one vHs crosses E_F for a given charge. This would additionally allow the experimental investigation of the many body instabilities expected in this configuration [8].

In this communication we report an analysis by Scanning Tunneling Microscopy (STM) and Spectroscopy (STS), complemented by tight binding calculations, of the density of states of electron doped twisted bilayers in a wide range of angles $(1.5^{\circ} < \theta < 15^{\circ})$ [9]. We have investigated the electronic structure of twisted bilayers (tBL) doped by charge transfer from the 6H-SiC(000-1) substrate (SiC-C face). Spectroscopic studies [10] have indeed revealed that few layers graphene films grown on this substrate are electron doped in the 10^{12} cm⁻² range. The doping level is larger for the plane closest to the substrate and decreases for the next layer. This charge distribution results in a different electric potential on each graphene plane [11], which shifts the electronic energy levels (e. g. the Dirac points) of the layers accordingly. We have selected tBL grown on the (3x3) reconstruction of the SiC-C face (Fig. (a)), for which the perturbation induced by the substrate is minimal [12]. Analyzing 17 independent tBL find by STS that the vHs (an example is shown in Fig. (b)) crosses the Fermi level for $\theta \approx 3^{\circ}$ for bilayer islands (Fig. (c)), with no change in the vHs splitting compared to the neutral case [4]. The layer dependent doping results in a difference in the

electrical potentials of the two layers of the order of 0.1 V at most, as deduced from a simple model supported by experimental observations.



Figure : Position of the upper vHs relative to the Fermi level as a function of the rotation angle. (a) STM image $(50 \times 50 \text{ nm}^2, \text{ sample bias: } -2.0 \text{ V})$ of a bilayer island with $\theta = 2.3^\circ$ (highlighted by the green square). (b) Local STS spectra acquired at topographic maxima (red curve) and minima (black curve) of the bilayer island in (a). Stabilization bias/current: +0.5 V/0.3 nA. The curves have been shifted vertically for clarity. The green (orange) arrows indicate the positions of the upper (lower) vHs at E_+ (E_-). E_+ is located below the Fermi level E_F for this value of θ . (c) Energy E_+ (relative to E_F) of the upper vHs as a function of θ .

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